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Introduction

- Lightning produces NO because the extreme temperatures (>20000 K) in lightning channels dissociate molecular O₂ and molecular N₂, which then combine to form NO, which quickly reacts with O₃ to form NO₂. Lightning is responsible for 10-15% of NO_x emissions globally. This is 2 – 8 Tg N a⁻¹ [Schumann and Huntrieser, 2007] or 100 to 400 mol per flash. Much of the uncertainty stems from limited knowledge of lightning NO_x production per flash (LNO_x PE) or per unit flash length.
- Most LNO_x is injected into mid- and upper-troposphere where away from deep convection its lifetime is longer relative to lower troposphere NO_x. NO_x in this region enhances the concentrations of upper tropospheric NO_y, OH, and O₃ and contributes to positive radiative forcing by O₃ and negative forcing by CH₄.
- We have previously used OMI NO₂ to obtain estimates of LNO_x production per flash over the Gulf of Mexico [Pickering et al., 2016, JGR], in convective events during NASA's TC4 field program [Bucsele et al., 2010, JGR], and over broad regions of the tropics [Allen et al., 2019, JGR] and midlatitudes [Bucsele et al., 2019, JGR]. In the latter studies, we obtained PE values of 170 ± 100 mol flash and 180 ± 100 mol flash, respectively.

TROPOMI LNO_x PE Algorithm

$$PE = [V_{\text{tropNO}_x} \times \Sigma \text{Area}] / [N_A \times \Sigma (\text{Flashes} \times \exp(-t / \tau))]$$

PE ≡ LNO_x Production Efficiency (moles NO_x/flash)

V_{tropNO_x} ≡ Median vertical column density (VCD) of LNO_x over pixels within ROI¹ that satisfy the DCC².

Area ≡ Area of pixels within ROI that satisfy the DCC² or have P < 500 hPa and undefined cloud-fractions

N_A ≡ Avogadro's Number

Flashes ≡ Number of GLM flashes⁵ within ROI during 5 hour period before TROPOMI overpass time⁶

t ≡ Age of individual flashes

τ ≡ Lifetime of NO_x in near field of convection

¹Region of interest (ROI) ≡ Latitude-longitude region encompassing deep convective system

²Deep convective constraint (DCC) ≡ Cloud fraction³ > 0.95 and cloud pressure⁴ < 500 hPa

³Cloud Fraction ≡ cloud_fraction_crb_nitrogen_dioxide_window from TROPOMI support data

⁴Cloud pressure ≡ cloud_pressure_crb from TROPOMI support data

⁵GLM flashes are not adjusted for detection efficiency or false alarms

⁶Overpass time ≡ Time TROPOMI exited ROI

$$V_{\text{tropNO}_x} = \text{Median}(V_{\text{tropNO}_x} - V_{\text{tropbkgn}})$$

$$V_{\text{tropNO}_x} = [S_{\text{NO}_2} - \text{avg}(V_{\text{stratNO}_2} \times \text{AMF}_{\text{strat}})] / \text{AMF}_{\text{LNO}_x} \quad [\text{avg over all pixels within ROI satisfying DCC}]$$

S_{NO₂} ≡ NO₂ Slant Column Density (SCD) for individual DCC pixels within ROI

V_{stratNO₂} ≡ Stratospheric VCD of NO₂ for DCC pixels within ROI

AMF_{strat} ≡ Stratospheric air mass factor for DCC pixels within ROI

AMF_{LNO_x} ≡ AMF converting tropospheric slant column of NO₂ to vertical column of LNO_x. Unless otherwise specified it is assumed to equal 0.46 following Beirle et al. (2009, ACP)

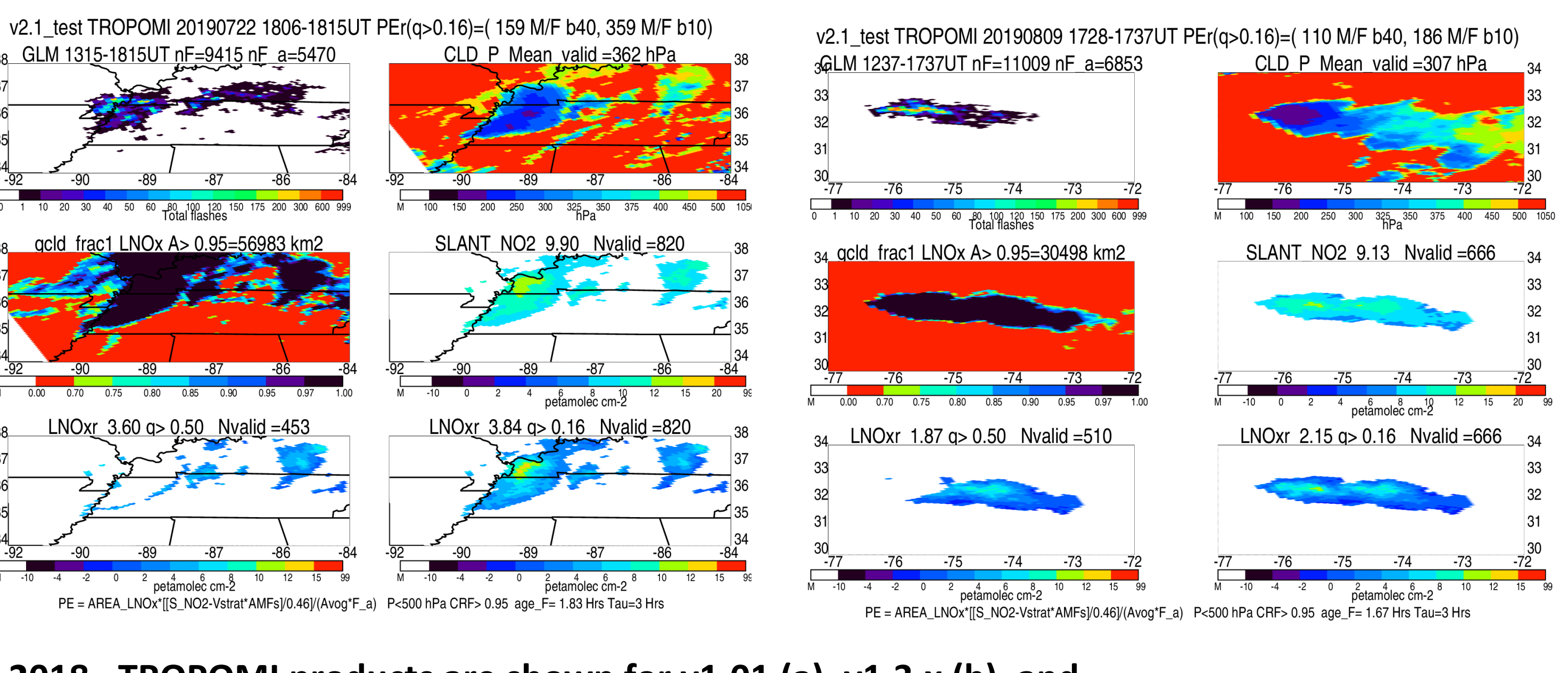
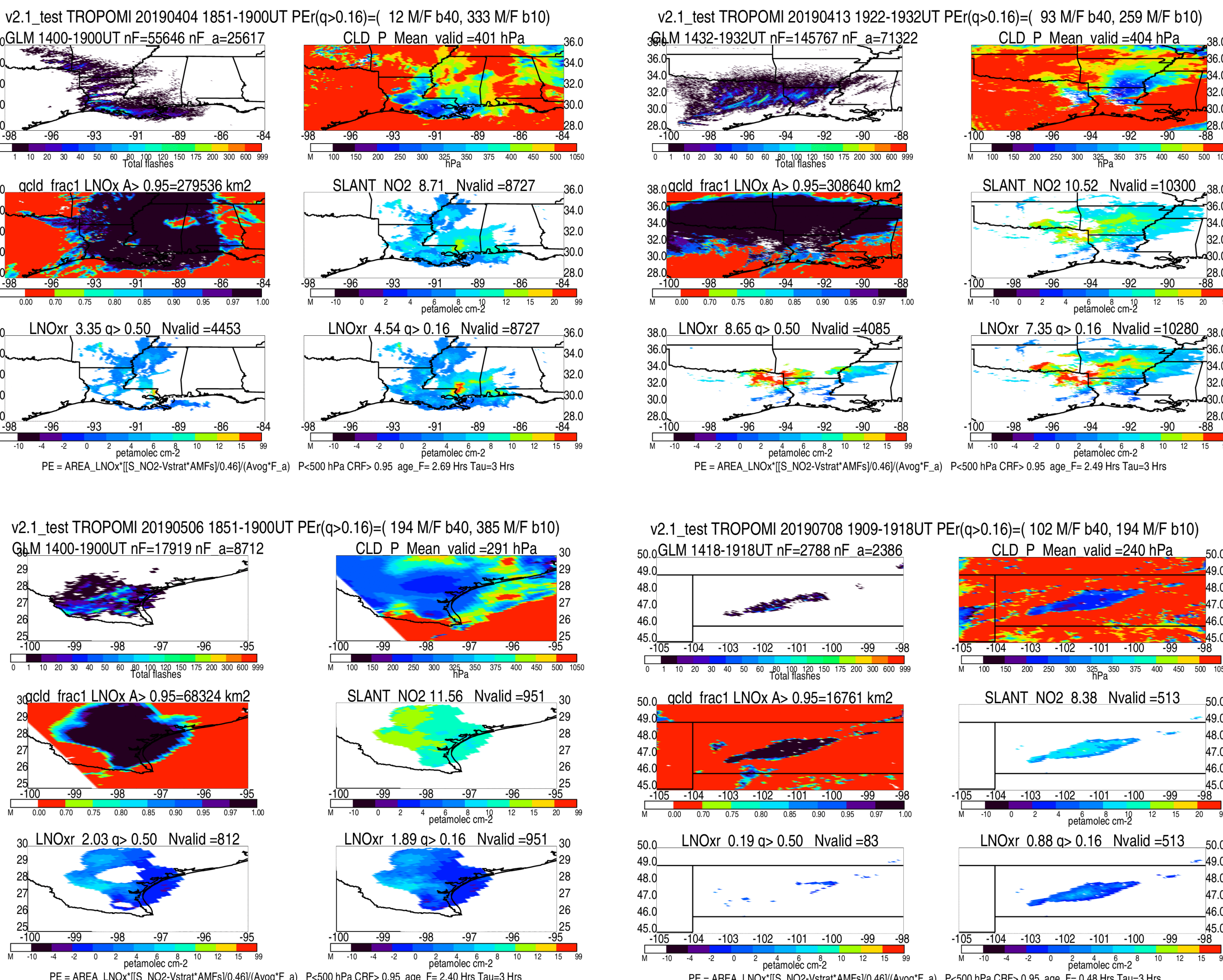
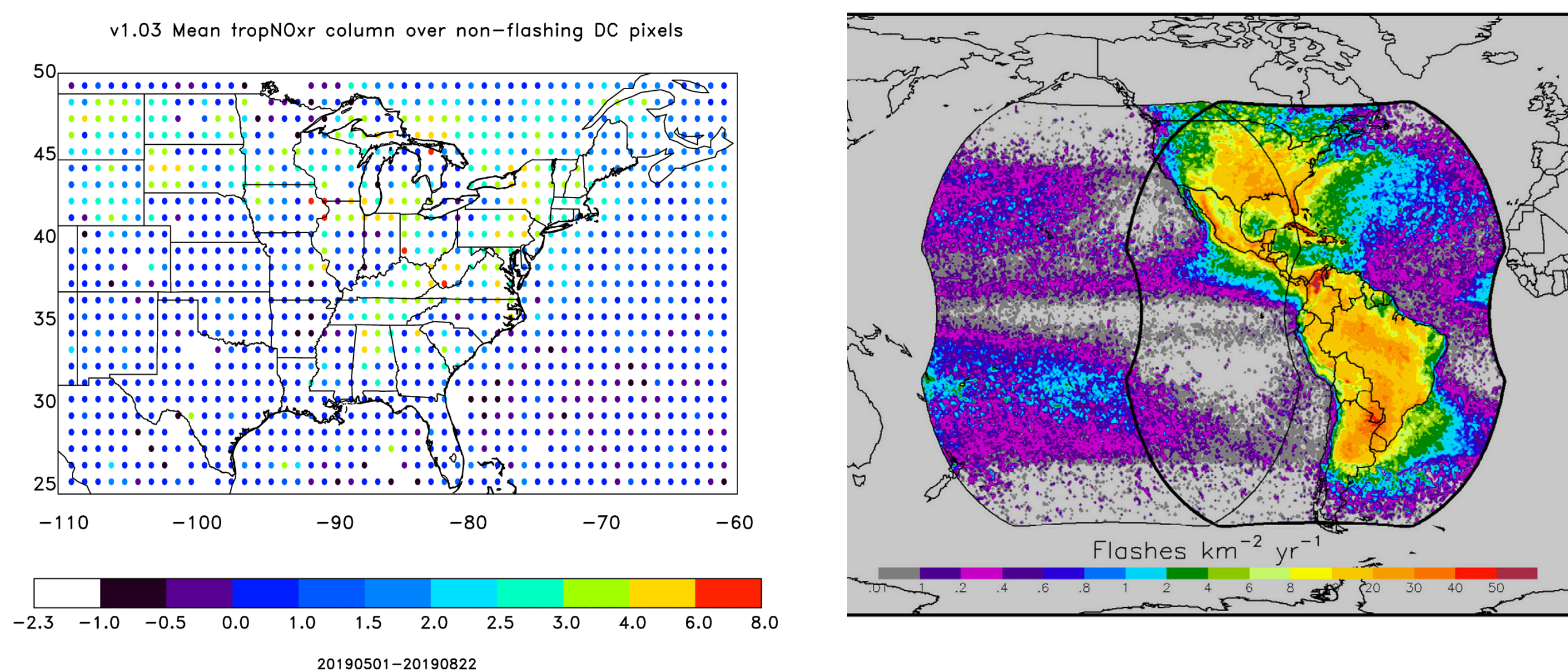
V_{tropbkgn} is estimated using 3 different methods

V_{tropbkgn10} (V_{tropbkgn10}) ≡ 10th (40th) % of V_{tropNO_x} for non-flashing pixels within ROI satisfying DCC.

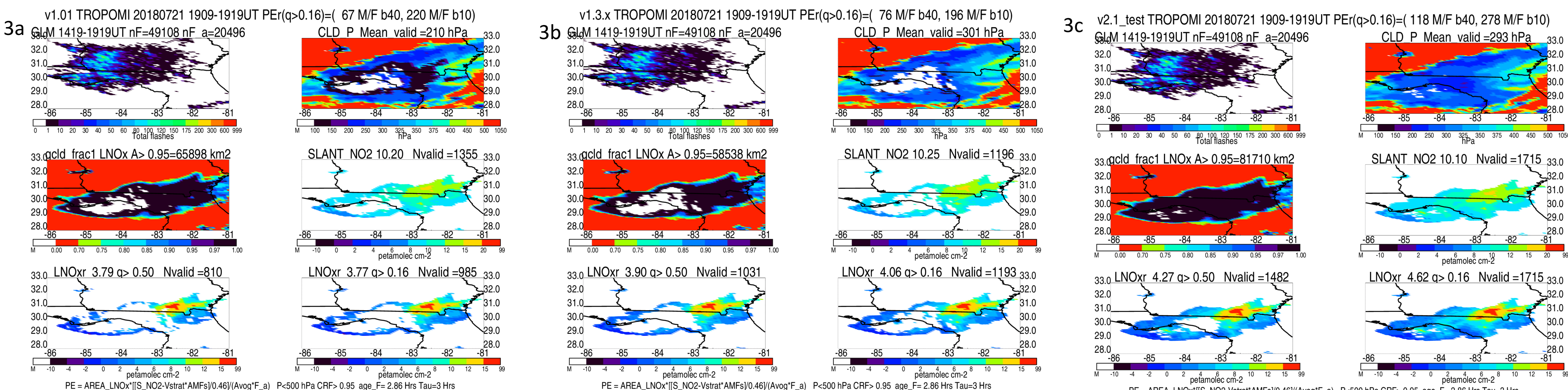
V_{tropbckl} ≡ Mean value of V_{tropNO_x} for pixels satisfying the DCC within ROI on low-flash days between May 1 & Aug 22, 2019

Fig. 1. V_{tropbckl}: Values obtained by applying 5° box car smoother to 1° × 1° gridded values of V_{tropNO_x}, obtained using pixels on low-flash days (< 10000 GLM flashes during 5-hour period preceding TROPOMI overpass) that are more than ~50 km distant from lightning.

Fig. 2. Idealized representation of GLM flashes Observed by GLM-17 (left region) and GLM-16 (right region)



Figures 3a-c show GLM and TROPOMI products over deep convection observed on July 21, 2018. TROPOMI products are shown for v1.01 (a), v1.3.x (b), and v2.1_test (c). See Figure 4 caption for details on individual plots. For this system over the panhandle of Florida on 21 July 2018, the number of valid good (fair) quality VLNO_x retrievals over pixels influenced by deep convection and/or lightning increased from 810 (985) in v1.01, to 1031 (1193) in v1.3.x, to 1482 (1715) in v2.1_test leading to more robust estimates of LNO_x PE.



TROPOMI

The data suite for the Tropospheric Monitoring Instrument (TROPOMI) [Veefkind et al., 2012] onboard the Copernicus Sentinel-5 Precursor satellite that began operating in its nominal mode in late April 2018 includes NO₂ and cloud products such as cloud fraction and cloud top pressure. The TROPOMI NO₂ processing system is an improved version of the KNMI DOMINO system that retrieves slant columns from Level 1b radiances using DOAS; 2) separates the tropospheric and stratospheric slant columns based on data from the TMS model and assimilation system [Huijnen et al., 2010]; and 3) converts the tropospheric and stratospheric slant columns to vertical columns by application of air mass factors (AMF) which include daily information on NO₂ vertical distributions from the TMS model at 1° × 1° resolution. The horizontal resolution of the NO₂ products at nadir are approximately 3.6 km (cross track) × 7.2 km (along-track) prior to August 6, 2019 and 3.6 × 5.6 km after August 6, 2019. The TROPOMI NO₂ retrieval uses cloud-pressures from the FRESCO-S algorithm, which is based on the FRESCO+ algorithm described in Wang et al. [2008]. Cloud fraction information is retrieved from the NO₂ spectral window and accounts for Rayleigh scattering.

This study uses TROPOMI products from TROPOMI v1.01, v1.3.x (processor version 1.03), and v2.01_test where the latter is a modified Copernicus Sentinel data product created for this study that includes spike removal and has better flagging for saturation and flagging for blooming allowing for increased data coverage over bright (flashing scenes). Overexposure of CCDs (saturation) is common for TROPOMI scenes affected by lightning. Blooming occurs when the influence of saturation spreads to neighboring wavelengths and pixels.

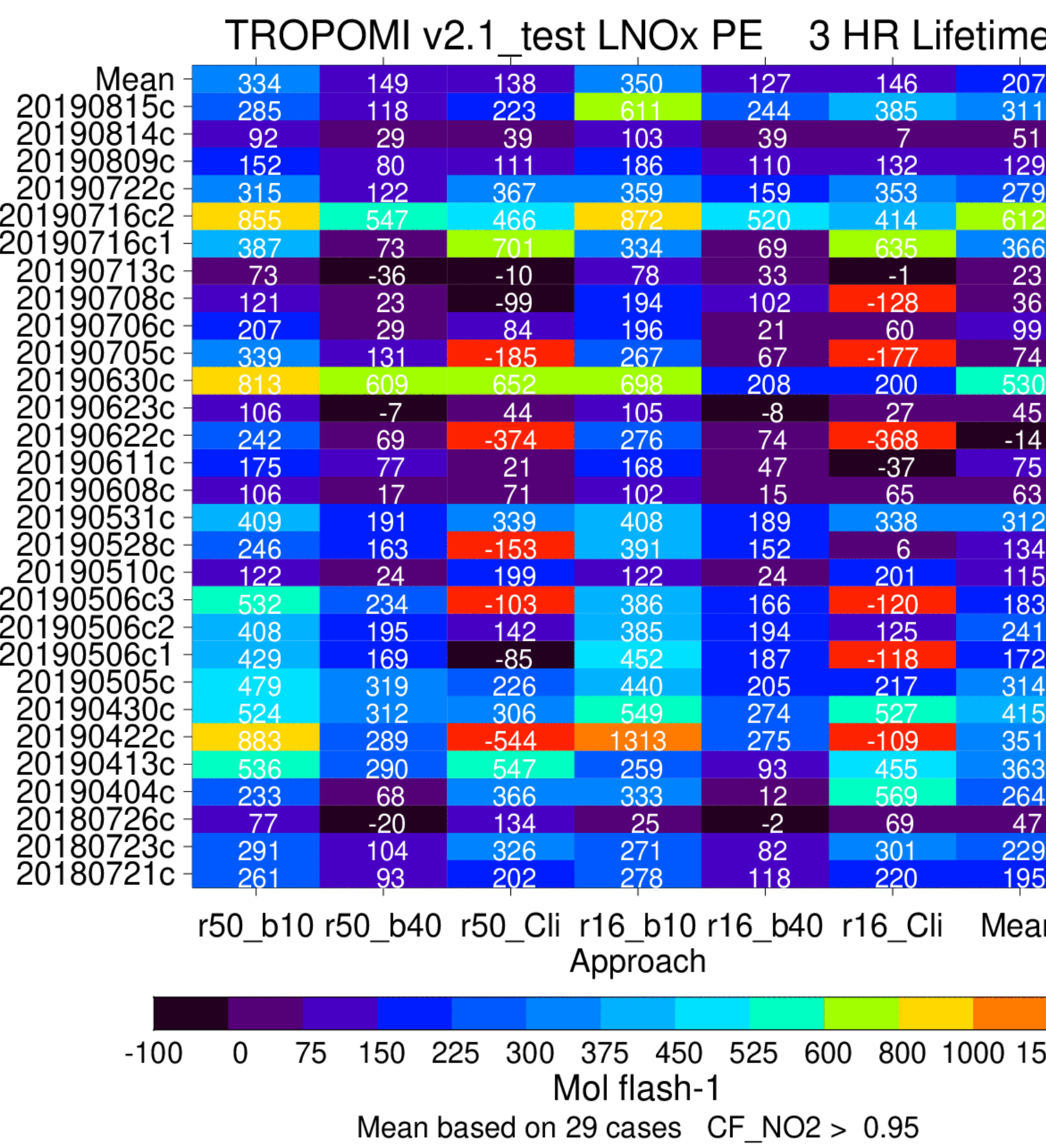
Table 1. Details on the 29 case studies used to estimate LNO_x PE from GLM16 flashes and v2.1_test TROPOMI data. The table lists the date and location of each convective systems as well as details needed to estimate the LNO_x PE from each case.

In this table, Area is given in km², the age of flashes (Age_Fl) is given in hours, VCDs of NO_x are given in peta mole cm⁻², and the PE is given in mol per flash. Nflashes gives the number of GLM flashes while Nflash_a is the number of flashes after adjusting for chemical decay assuming a chemical lifetime of 3 hours. Negative values of PE indicate that background columns over non-flashing grid boxes exceed the median columns in the region.

YYYYMMDD	Region	Area	Nflashes	Age_Fl	Nflash_a	VLNOxr	VLNOxb30	VLNOxb02	VLNOxbcl1	LNOxPE40	LNOxPE10	LNOxPECl1	NPTS
20180721c	86W-81W 28N-33N	81710	49108	2.86	20496	4.62	2.84	0.42	1.29	117	277	220	1715
20180723c	90W-82W 24N-30N	122176	117126	2.53	56090	8.57	6.3	1.09	0.25	81	270	300	2738
20180726c	83W-79W 28N-31N	27874	23194	1.83	13516	2.35	2.41	1.6	0.33	-2	25	69	370
20190404c	98W-84W 28N-36N	279536	55646	2.69	25616	4.54	4.48	2.71	1.4	11	332	569	8727
20190413c	100W-88W 28N-38N	308640	145767	2.49	71322	7.35	6.05	3.74	1.01	92	259	454	10280
20190422c	98W-88W 38N-48N	22733	2479	3.85	734	1.67	1.14	-0.88	1.89	275	1312	-109	447
20190430c	105W-87W 33N-45N	287191	49351	2.39	24997	4.22	2.79	1.34	1.46	274	549	526	8101
20190505c	86W-78W 25N-31N	109140	40486	2	22908	3.14	0.55	-2.42	0.4	204	440	217	1733
20190506c1	102W-95W 38N-43N	49985	4247	2.39	2076	1.21	0.74	0.08	1.5	186	451	-118	1027
20190506c2	100W-95W 25N-31N	68324	17919	2.4	8711	1.89	0.4	-1.06	0.93	193	384	125	951
20190506c3	82W-73W 25N-31N	55903	9950	1.42	6519	-0.34	-1.51	-3.05	0.5	166	386	-120	1092
20190510c	98W-91W 25N-30N	60569	58727	2.98	24140	6.13	5.54	3.19	1.32	24	122	200	2173
20190528c	98W-88W 37N-43N	107665	21100	2.07	11850	1.88	0.87	-0.72	1.84	151	391	6	2578
20190531c	70W-60W 35N-40N	44355	19662	3.17	7346	4.2	2.31	0.14	0.83	189	407	337	845
20190608c	90W-78W 25N-35N	120395	56841	2.13	31535	2.14	1.9	0.53	1.12	15	101	64	3901
20190611c	87W-78W 25N-31N	45181	16586	1.36	11192	-0.14	-0.84	-2.65	0.42	47	168	-37	625
20190622c	102W-96W 40N-46N	31459	5982	2.72	2634	0.02	-0.17	-1.19	2.06	73	275	-367	999
20190623c	98W-88W 33N-38N	124840	41602	2.34	21357	1.22	1.3	0.14	0.94	-7	105	26	3070
20190630c	95W-87W 42N-48N	146174	27135	2.78	12118	2.94	1.9	-0.55	1.94	208	697	199	3250
20190705c	99W-92W 36N-41N	88902	20135	2.4	10179	0.19	-0.27	-1.65	1.42	67	266	-177	1191
20190706c	88W-82W 27N-31N	74759	21018	1.9	12049	1.4	1.2	-0.5	0.83	21	196	59	1038
20190708c	105W-98W 45N-50N	16761	2788	0.48	2386	0.88	0.01	-0.78	1.98	102	193	-128	513
20190713c	83W-77W 33N-36N	52490	32998	1.12	23474	1.7	0.82	-0.4	1.73	32	77	0	788
20190716c1	96W-92W 41N-45N	19278	3558	2.61	1664	5.48	5.12	3.74	2.18	69	333	634	282
20190716c2	94W-88W 31N-36N	31520	6777	3.25	2623	3.15	0.54	-1.22	1.07	519	872	414	483
20190722c	92W-84W 34N-38N	56983	9415	1.83	5469	3.84	2.93	1.77	1.81	158	359	352	820
20190809c	97W-72W 30N-34N	30498	11009	1.67	6853	2.15	0.66	-0.36	0.36	110	185	132	666
20190814c	92W-82W 28N-33N	82472	50337	1.4	33725	1.78	0.83	-0.75	1.61	38	102	6	3021
20190815c	100W-92W 38N-43N	56608	9857	3.09	3861	3.27	2.27	0.76	1.69	244	610	385	2191

Below:

LNO_x PE for each of the 29 cases (y-axis) as derived using v2.1_test TROPOMI products and assuming a 3-hour lifetime. Over the x-axis, r50 (r16) indicates that only pixels with quality flags > 0.50 (0.16) are used in estimating PE. The suffixes b10 and b40 indicate that the tropospheric background is day-specific and given by the 10th (40th)% column over non-flashing grid boxes. The suffix Cli indicates that the climatological mean background for that location is used.



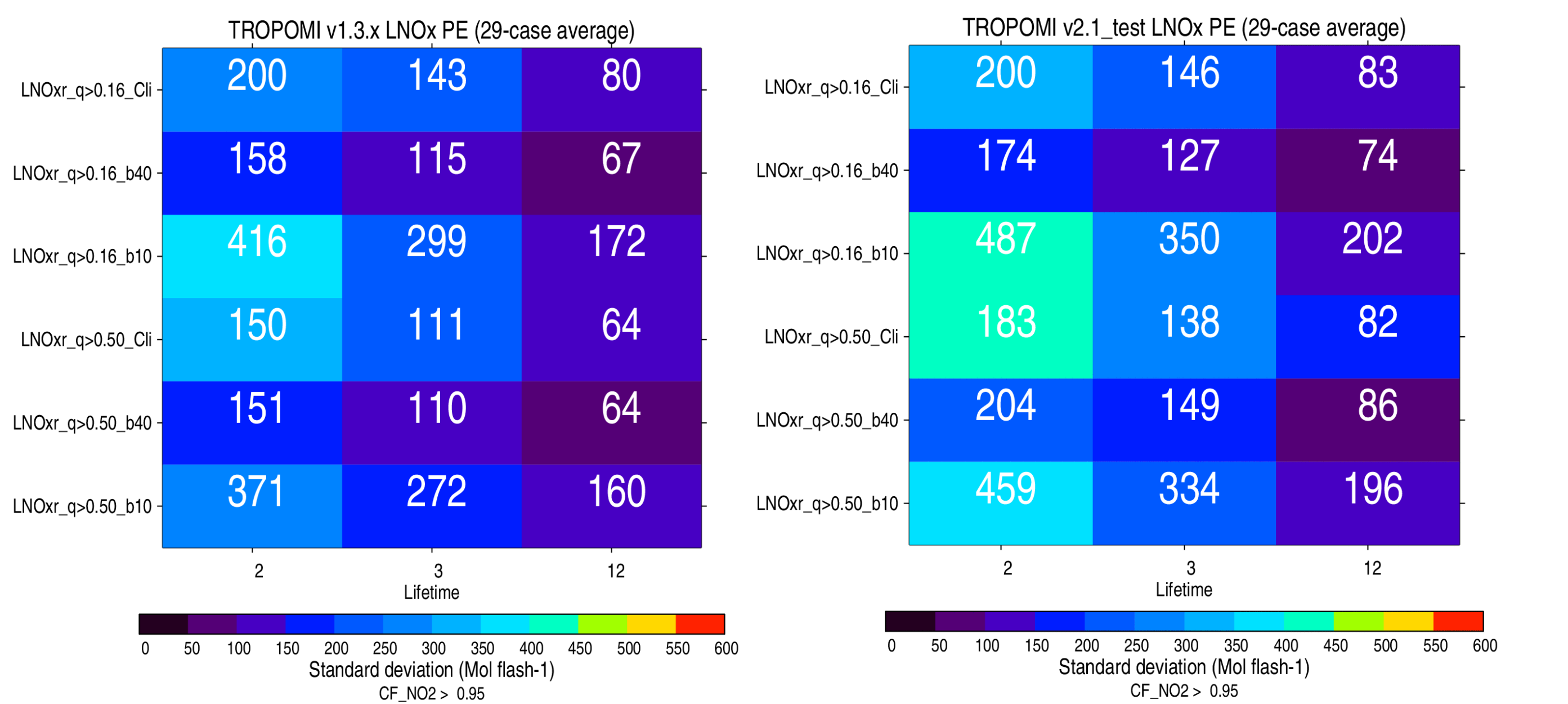
Below:

LNO_x PE as a function of chemical lifetime of NO_x (2, 3, or 12 hours) for 3 different methods of estimating the tropospheric background (b10, b40, and Cli), two pixel quality choices (Quality flag > 0.16 or 0.50), and 2 versions of TROPOMI retrievals (TROPOMI v1.3.x and TROPOMI v2.1_test). Colors show the standard deviations for the 29 cases.

For a 3-hour lifetime, LNO_x PE = 299 (350) mol per flash when calculated using v1.3.x (v2.1_test) TROPOMI pixels with quality flags exceeding 0.16 when the 10th% VCD over non-flashing pixels on the day-of interest is used in estimating the tropospheric background.

For a 3-hour lifetime, LNO_x PE = 115 (127) mol per flash when calculated using v1.3.x (v2.1_test) TROPOMI pixels with quality flags exceeding 0.16 when the 40th% VCD is used in estimating the background.

For a 3-hour lifetime, LNO_x PE = 143 (146) mol per flash when calculated using v1.3.x (v2.1_test) TROPOMI pixels with quality flags exceeding 0.16 when the climatological background is used for the background.



Uncertainties

- The AMF used to convert SCDs of NO₂ to VCDs of NO_x is assumed to equal 0.46. However, in reality it varies with viewing geometry, Rayleigh and Mie scattering, the vertical profile of NO₂, and the NO / NO₂ ratio within a deep convective system (e.g., Silvern et al., 2018).
- The lifetime of NO_x in the near field of convection varies from 2-12 hours depending on the proximity to deep convection (e.g., Nault et al. (2016)).
- LNO_x PE is sensitive to the VCD of NO_x due to sources other than recent lightning (e.g., Allen et al., 2019).
- GLM flashes are not adjusted for detection efficiency or for false alarms.
- TROPOMI columns are often missing over bright regions where flashes and presumably VLNO_x are large.

Summary

- LNO_x PE was estimated using GLM flashes and TROPOMI NO₂ columns for 29 convective systems observed during the spring- and summer of 2018-2019
- LNO_x PE varied greatly from case-to-case with the mean PE for a 3-hour lifetime ranging from 110 to 350 mol per flash depending on TROPOMI version, background approach, and quality flag thresholds.
- Tropospheric NO₂ retrievals with TROPOMI are difficult over deep convective scenes due to small tropospheric AMFs, saturation of CCD pixels affected by lightning and blooming effects. However, tweaks to the processing algorithm allow more retrievals over these scenes.
- Future work will include the incorporation of case-specific AMFs, refinement of the tropospheric background approach, and analysis of the representativeness of these 29 cases.